Structural phase transitions induced in Ni_2MnGa by low-temperature uniaxial compression

A. N. Vasil'ev and A. Kaïper
M. V. Lomonosov Moscow State University, 119899 Moscow, Russia

V. V. Kokorin and V. A. Chernenko
Institute of Metal Physics, Ukrainian Academy of Sciences, 252680 Kiev, The Ukraine

T. Takagi and J. Tani
Institute of Fluid Science, Tohoku University, Sendai 980, Japan

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The temperature dependence of the low-field magnetic susceptibility, the specific heat, and the transverse sound velocity has been measured in single crystals of the ferromagnet Ni_2MnGa compressed along the $\langle 110\rangle$ axis. The relative deformation was $\Delta L/L=0.05$. The load was removed at liquid-nitrogen temperature. The dimensions and shape of the crystals are restored upon heating as the result of a sequence of first-order structural phase transitions accompanied by clearly defined features on the curves of the physical properties studied. The new phases appear in Ni_2MnGa because of an anomalous instability of the crystal lattice of this compound with respect to atomic displacements along a $\langle 110\rangle$ direction in the (110) plane.

The ferromagnetic intermetallic compound Ni_2MnGa is distinguished from other Heusler alloys in that when cooled below room temperature it undergoes a structural transition from a cubic $L2_1$ α phase to a tetragonal β_1 phase with an axis ratio $c/a=0.94$. The very first studies[1] of this compound by neutron diffraction revealed some additional reflections, due to long-period modulations of the crystal lattice in the β_1 phase. Subsequent measurements by x-ray diffraction dilatometry[2,3] showed that the tetragonal lattice of Ni_2MnGa is modulated by a static transverse displacement wave with a wavelength equal to five lattice constants. The wave vector of this wave is perpendicular to (110) planes, and the polarization vector is along a $\langle 110\rangle$ crystallographic direction. The period of this static displacement wave can be varied at low temperatures by compressing the material along the $\langle 110\rangle$ direction. As the load is increased, the five-layer modulation of the original phase gives way to a seven-layer modulation, and then Ni_2MnGa goes into an unmodulated tetragonal phase. Since the structural transitions in this compound have hysteresis, occur at low temperatures, and are accompanied by small changes in volume, the compound Ni_2MnGa clearly exhibits shape memory and superplasticity. Furthermore, since the structural transitions in Ni_2MnGa occur in a ferromagnetic matrix, the dimensions and shape of this material can be controlled by using some combination of the temperature, a magnetic field, and a uniaxial compression.

The first steps toward a realization of this program were taken in the study which we are reporting here. We measured several physical properties of the phases which

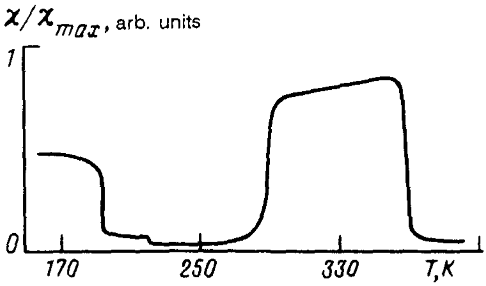

FIG. 1. Temperature dependence of the low-field magnetic susceptibility χ of a Ni_2MnGa single crystal compressed 5% along the ⟨110⟩ direction and unloaded at $T=77$ K.

are induced by low-temperature compression of Ni_2MnGa single crystals. The low-field magnetic susceptibility χ, the specific heat C, and the transverse sound velocity S were measured on samples compressed along the ⟨110⟩ direction and again after the load was withdrawn at liquid-nitrogen temperature. The maximum relative deformation $\Delta L/L$ at which no plastic flow was observed was about 5%. The maximum value was reached at a pressure on the order of 100 MPa. To avoid going beyond this limit, we placed the sample in a confining ring of heat-treated beryllium bronze in a low-temperature press.

This formulation of the problem assumes contactless measurements, since the state of the material is sensitive to even small stresses over the entire temperature range, especially at the temperatures of phase transitions. While there is no difficulty in contactless measurements of χ and C—in the first case, the sample is positioned in an inductance coil, and in the second it is put in a thermal-analysis chamber—in the case of S the standard acoustic methods are unsuitable. Aside from the fact that a substantial stress always arises at a point where the sample makes contact with the piezoelectric transducer, and this stress varies with the temperature, it is simply not possible to achieve acoustic contact at liquid-nitrogen temperature capable of withstanding large changes in the dimensions and shape of the sample upon heating. Accordingly, to measure the temperature dependence of the transverse sound velocity we used a procedure involving contactless excitation and detection of elastic waves. This procedure is based on the direct conversion of electromagnetic and acoustic waves at the boundary of a metal in an external magnetic field.[4] During the heating, we observed some anomalies on the temperature dependence of the physical properties which we measured. These anomalies correspond to the reverse sequence of phase transitions: from the unmodulated tetragonal β_3 phase to the tetragonal β_2 phase with the seven-layer modulation ($T_3 \sim 190$ K) to the tetragonal β_1 phase with a five-layer modulation ($T_2 \sim 230$ K) to the cubic α phase ($T_1 \sim 290$ K). The phase-transition temperatures found from the various physical measurements agree within 5°. The scatter in their values is due primarily to the extreme sensitivity of Haeusler alloys to the stoichiometry; even slight variations in the composition lead to large shifts of the phase-transition temperatures. For example, Curie points T_C from 362 to 395 K have been reported in the literature[5] for Ni_2MnGa.

Figure 1 shows the temperature dependence of the low-field magnetic susceptibility χ. The β_3 phase has the largest value of χ, comparable to the susceptibility of the

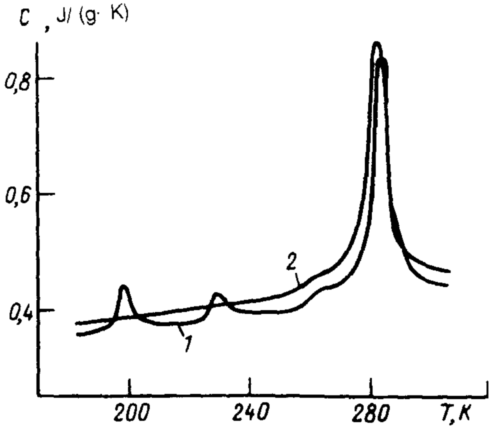

FIG. 2. *1*—Temperature dependence of the specific heat C of a Ni_2MnGa single crystal compressed 5% along the ⟨110⟩ direction and unloaded at $T=77$ K; 2—the same, for an unloaded sample.

high-temperature α phase. The reason for the sharp decrease in χ at 190–290 K is that the static displacement wave along one of the ⟨110⟩ directions creates a magnetic anisotropy in the β_2 and β_1 phases. The magnitude of this anisotropy is smaller in the β_2 phase, with the seven-layer modulation. The body-centered tetragonal β_3 phase and the body-centered cubic α phase do not have this one-dimensional modulation. The decrease in χ at $T_c=370$ K corresponds to a transition of the α phase to a paramagnetic state.

Curve *1* in Fig. 2 shows the temperature dependence of the specific heat of a Ni_2MnGa sample compressed and unloaded at liquid-nitrogen temperature. During the heating we also observed the sequence of structural transitions described above. The heat of the transitions from the β_3 phase to the β_2 phase (0.37 J/g) and from β_2 to β_1(0.175 J/g) is an order of magnitude smaller than the heat of the transition from β_1 to α. The clearly expressed heat of transitions indicates that all these structural conversions in Ni_2MnGa are first-order phase transitions. Curve *2* in Fig. 2 shows the temperature dependence found for the specific heat as the unloaded sample was cooled. In this formulation of the experiment, there are no peaks reflecting low-temperature structural conversions.

Figure 3 shows the temperature dependence of the transverse sound velocity S in a Ni_2MnGa single crystal; the sound is propagating along the $\langle 1\bar{1}0\rangle$ direction and is polarized along the ⟨110⟩ direction. In this case the sound wave vector is perpendicular to the compression axis, while the polarization vector coincides with that axis. According to Figs. 1 and 2, the jumps in S in Fig. 3 correspond to transitions from the β_3 phase to the β_2 phase ($T_3=190$ K) and from β_2 to β_1($T_2=230$ K). Since the β_3 phase has an unmodulated tetragonal lattice, and its ⟨110⟩ crystallographic axis coincides with the corresponding ⟨110⟩ axis in the α phase, the measured velocities can be related to the magnitude of the elastic modulus $(C_{11}-C_{12})/2$, which characterizes the elasticity of the lattice with respect to a displacement along a ⟨110⟩ direction in the (110) plane. This approach can be extended in a qualitative sense to the unmodulated tetragonal phases. We see that the stability of the lattice with respect to shear stress increases as we go from β_3 to β_2 and from β_2 to β_1. On the temperature

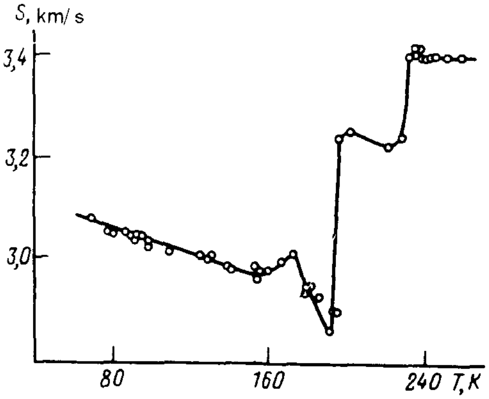

FIG. 3. Temperature dependence of the transverse sound velocity S in a Ni_2MnGa single crystal compressed 5% along the ⟨110⟩ axis and unloaded at $T=77$ K.

dependence of the transverse sound velocity and of the specific heat we can see some clearly defined anomalies at the structural transitions. We also see some features at other temperatures; the nature of these other features is not clear at this point.

This study of the physical properties of the phases induced by low-temperature uniaxial compression of Ni_2MnGa single crystals has revealed that the restoration of the shape and dimensions of this material, when the load is removed and when the material is then heated, occurs because of first-order structural phase transitions at clearly defined temperatures. The appearance of new phases in Ni_2MnGa results from the anomalous instability of the crystal lattice of this compound with respect to atomic displacements in the ⟨110⟩ direction in the (110) plane. The intimate relationship between the magnetic and elastic properties in Ni_2MnGa, in particular, the substantial increase in the magnetic anisotropy in the modulated tetragonal phases in comparison with that in the unmodulated tetragonal and cubic phases, opens up the possibility of using a magnetic field to control the shape and dimensions of these crystals.

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